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MEASUREMENTS OF AEROSOL SIZE DISTRIBUTIONS IN THE LOWER TROPOSPHERE OVER NORTHERN EUROPE

Bruce W. Fitch
Ted S. Cress

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June 1981

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Optical Physics Division

Prepared for
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Airborne measurements of particle size distributions were made at several altitudes within and above the mixing layer at sites near Ahlhorn and Meppen (W. Germany), Rodby (Denmark) and Bruz (France). The distributions were measured over the range 0.2 to 5.9 micrometers (μm) in particle radius using a Royco model 220 particle counter. The experimental data gathered at the sites were analyzed in terms of volume-size distributions instead of the commonly used particle number distributions. The volume distribution plots were found to be

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bimodal with the accumulation mode centered at a mean particle radius in the range 0.26 to $0.49\mu m$ and the coarse particle mode existing beyond $1.0\mu m$. The data show the accumulation mode is well defined by a log-normal distribution. The values of the measured volume scattering coefficient and mean particle radius of the accumulation mode increase as the maximum particle volume of the mode increases. The existence of an accumulation mode was almost always confined to the mixing layer. It is interesting that haze layers above the mixing layer were found to have a distinct coarse particle mode but, generally, no distinct accumulation mode. The total concentration of particles in each of the two modes appear related for larger values of concentration in the coarse particle mode.

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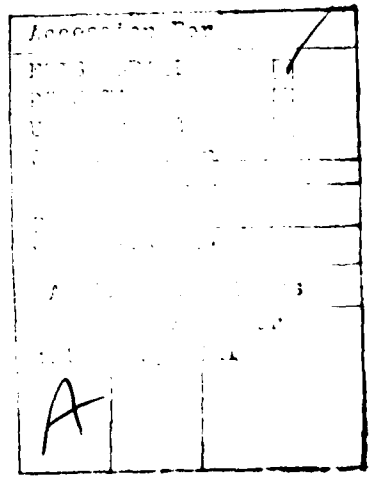

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AIR FORCE GEOPHYSICS LABORATORY
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UNITED STATES AIR FORCE
HANSCOM AFB, MASSACHUSETTS 01731

Summary

Airborne measurements of particle size distributions were made at several altitudes within and above the mixing layer at sites near Ahlhorn and Meppen (W. Germany), Rodby (Denmark) and Bruz (France). The distributions were measured over the range 0.2 to 5.9 micrometers (μm) in particle radius using a Royco model 220 particle counter. The experimental data gathered at the sites were analyzed in terms of volume-size distributions instead of the commonly used particle number distributions. The volume distribution plots were found to be bimodal with the accumulation mode centered at a mean particle radius in the range 0.26 to 0.49μm and the coarse particle mode existing beyond 1.0μm. The data show the accumulation mode is well defined by a log-normal distribution. The values of the measured volume scattering coefficient and mean particle radius of the accumulation mode increase as the maximum particle volume of the mode increases. The existence of an accumulation mode was almost always confined to the mixing layer. It is interesting that haze layers above the mixing layer were found to have a distinct coarse particle mode but, generally, no distinct accumulation mode. The total concentration of particles in each of the two modes appear related for larger values of concentration in the coarse particle mode.

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1. Introduction

Sophisticated numerical techniques have been developed by a number of investigators, including Dave (1978) and Takashima (1975), to solve for the radiation emerging from an atmosphere. These investigators and others, however, must in some way rely upon a knowledge of the size distribution of the atmospheric aerosol as a function of altitude, season, and location. The size distributions frequently used are the haze models of Deirmendjian (1969), who used a modified gamma function; the power-law size distribution function of Junge (1963) or a log-normal distribution as described by Aitchison and Brown (1957). Generally, the shape of the distribution is held constant while only the particle concentration changes with altitude.

There are few measurements to support the use of these model size distributions. A series of airborne measurements were made by Blifford and Ringer (1969), who collected aerosol samples in the central United States during 1966 at altitudes from about 0.3 to 9 km above the ground. A majority of the samples, 22 of them, were acquired near Scottsbluff, Nebraska. Sampling of the troposphere has also been conducted by Laulainen, Alkezweeny and Thorp (1978). To augment these data and increase our understanding of the atmospheric aerosol content, an extensive measurement program was conducted in the lower troposphere at seven sites in Northern Europe and Great Britain during each of the four seasons. This report presents the results of analysis of some of these data.

2. Method of Measurement

Aircraft measurements of the number and size distribution of the atmospheric aerosol were conducted along tracks of constant altitude above the ground near Bruz (France), Rodby (Denmark), Ahlhorn and Meppen (W. Germany) during the Spring and Fall of 1976 and the Summer of 1977. All of the 50 km long sampling tracks, the locations of which are shown in Fig. 1, were over land, except for the Rodby track which was over the Baltic Sea. These airborne aerosol measurements, described by Cress (1980), were part of a larger atmospheric optical measurement program (Duntley *et al.*, 1978) conducted under the sponsorship of the Air Force Geophysics Laboratory.

Sampling was performed in cloud free air at altitudes from about 150 to 6100 meters(m) above the surface. The altitudes varied from one flight to another because of changes in cloud height and mixing layer depth. Whenever possible, two samples were made at different altitudes within the mixing layer and two more above. The mixing layer was generally capped by an inversion layer. The sampling time of the Royco was typically four minutes, but occasionally samples were taken for one minute or as long as ten minutes, depending upon the situation and the density of particles being counted. A single flight required from 30 minutes to 3 hours and 40 minutes depending upon the number of altitudes flown.

The instrumentation on the C-130 aircraft included a Royco 220 single particle counter and an integrating nephelometer. Sample air for the Royco was provided by

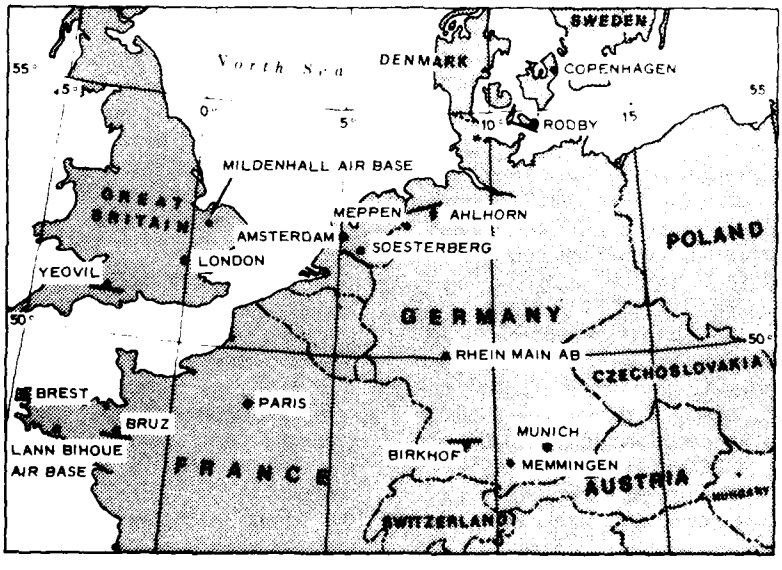

Fig. 1. Location of the Ground Base Data Sites (·) and the Aircraft Tracks(-).

a ram air isokinetic sampling probe located on top of the C-130 fuselage with the probe entrance 33 cms above the aircraft skin and 45 cms to the right of the radome which housed the nephelometer. The entire Royco system consisted of the Royco counter, a Technical Measurement Corporation Model 102 Gamma Scope II Pulse Height Analyzer, and a Hewlett-Packard Clock/Printer. The electronics of the analyzer were adjusted to position the counting of $0.4\mu m$ radius size particles in channel 18 of the total 89 channel range covering particle sizes 0.2 to $5.9\mu m$.

The Royco is a right-angle scattering counter that was calibrated with submicrometer size polystyrene spheres. The operating characteristics of this instrument have been the subject of a number of investigations including those of Quenzel (1969), Liu *et al.* (1974), and Cooke and Kerker (1975). The two fundamental sources of error result from an inadequate knowledge of the index of refraction of the ambient aerosol and modifications to the particle size distribution which may occur in the plumbing between the ambient air and the counter.

Investigations by Cooke and Kerker (1975) and Quenzel (1969) have demonstrated that significant errors in the determination of particle size can occur when the refractive index, m, of the particles is not known. Assuming a calibration with polystyrene-latex spheres (m=1.58-0i), Quenzel (1969) found that a maximum sizing error of a factor of 2.4 could occur when measuring carbon particles (m=1.95-0.66i) over the range of particle radius 0.1 to $3.0\mu m$. If the refractive index of water (m=1.33-0i) or an average atmospheric aerosol (m=1.50-0i) is used, the sizing error is less than a factor of 2.0. A value of m=1.50-0.02i, that includes absorption, produced a sizing error that increased with increasing particle radius. The magnitude of the error was difficult to determine from Fig. 2 of Quenzel (1969) but its maximum is estimated to be in the range 2 to 3. A ground based intercomparison study between a Royco 220 and active and classical Particle Measurement Systems aerosol counters showed agreement to within a factor of 2 to 5 in particle concentration (Cress and Fenn, 1978).

The physical properties of the ambient aerosol sample may have been modified in its passage from outside the aircraft to the Royco scattering chamber. Comparison of consecutive one minute data samples, however, indicate that the repeatability of the airborne Royco was good. The flow rate was approximately 47 cubic centimeters per second. Possible modifications to the aerosol sample may be a result of a drying of the particles or a selective loss of particles. The small number of changes in the direction of flow within the plumbing probably minimized the effects of particle loss. This is supported by the good agreement between this data set and the impactor data of Blifford and Ringer (1969) as shown by Cress (1980). The aerosol measurements most likely represent a low humidity or dry particle distribution because some evaporation probably occurred in the plumbing. Due to the small size range measured the shape of the distribution was probably preserved. These

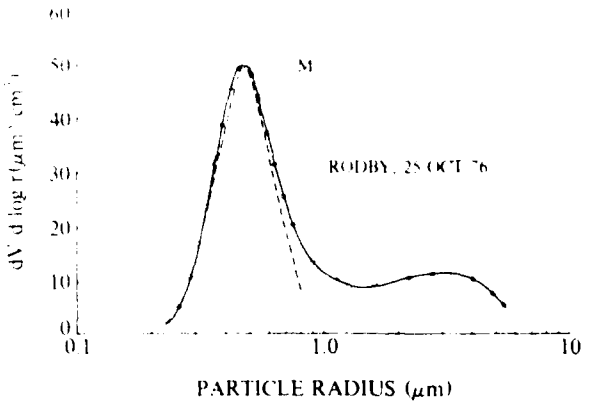

Fig. 2 The Aerosol Volume per Increment *dlogr* as a Function of Particle Radius for Data Taken at Rodby (Denmark) on October 25, 1976 at an Altitude of 305m above the Sea Surface (solid line). The mode radius, $\bar{r}$, and maximum, M_v, of the volume distribution accumulation mode are shown. A log-normal distribution with $\bar{r} = 0.45\mu m$ and standard deviation $\sigma = 0.13$ is shown (dashed line) for comparison.

sources of error are discussed in greater detail by Cress (1980).

The integrating nephelometer, described by Duntley *et al.* (1978), measures the volume scattering coefficient, s, over the range of scattering angle 5° to 170° and the scattering coefficient at 30° and 150°. The nephelometer operated at the photopic and in narrow wavelength bands centered at 0.478, 0.664 and $0.765\mu m$. Calibration using dry nitrogen and comparisons with an Eltro transmissometer indicate that the accuracy of the volume scattering coefficient is within 20% (Duntley *et al.*, 1976).

3. Results of Measurements

Traditionally, size distribution data have been presented in figures of log $(dN/dlogr)$ versus $logr$ where r is the particle radius and N is the total number density of particles smaller than r. A presentation of this type has been found to mask the multi-mode nature of distributions that appear when the data are plotted as volume (or mass) versus particle size, as pointed out by Whitby *et al.*, (1972) and Shettle (1975). For this reason it was decided to use the volume size display in this analysis.

A plot of $dV/dlogr$ versus $logr$ for the measurements taken at Rodby on October 25, 1976 at an altitude of 305m above the sea surface is shown in Fig. 2. Values of the total particulate volume, $dV/dlogr$, over an increment $dlogr$ are shown in the figure by circles for a few of the 89 data channels. The solid curve represents a fit to the data of all the channels. The inversion height was about 700m in this case as determined from temperature profile measurements made concurrently with the Royco measurements and from estimates of the surface haze layer thickness made by an on-board observer. The curve, shown in Fig. 2 centered about the radius $0.45\mu m$, is closely approximated by a log-normal

curve (dashed line). The symmetry and shape of the data curve is typical of the analyzed data. Whitby (1978) termed this the accumulation mode and stated that its main source of mass is from the coagulation of the smaller size aerosols and from gas to particle conversion. The peak in the curve at about $3\mu m$ indicates the presence of the coarse particle mode, which according to Whitby (1978) generally results from mechanical processes such as those that create wind blown dust or sea spray.

(a) ACCUMULATION MODE

For this study the accumulation mode is modeled by the value of the mode radius, $\bar{r}$, the standard deviation, σ, and by M_v - the value of $dV/dlogr$ occurring at the mode radius. Values of M_v are plotted against those of $\bar{r}$ in Fig. 3 for all of the seasons, stations, and altitudes below the inversion height that were available for this study. Values of M_v greater than $40\ \mu m^3/cm^3$ represent data collected only at Rodby during the winter. The figure shows values of M_v increasing with values of $\bar{r}$. Increasing values of M_v do not necessarily imply, however, that the particle number is increasing. Also shown in the figure are curves of $dV/dlogr$ for constant particle number, $dN/dlogr$, which illustrates the r^3 dependence inherent in the volume-size distribution. For values of $\bar{r}$ less than $0.34\mu m$ there is an increase in the measured particle number with increasing $\bar{r}$. For values of $\bar{r}$ greater than $0.34\mu m$, however, the particle number appears to be constant with increasing values of $\bar{r}$. The data presented in Fig. 3 suggest that there is a functional relationship between $\bar{r}$ and M_v. This relationship will be more fully explored for different air mass types using the complete set of Royco measurements. Plots comparing σ, which range in value from 0.08 to 0.20, with values of both M_v and total particle volume, V, of the mode show no discernible relationship between these parameters. This is probably due to the sensitivity of the determination of σ to the fitting procedure. In order to more easily visualize the character of the accumulation mode and to simplify its log-normal description the value $M_v = V/\sigma(2\pi)^{\frac{1}{2}}$ is used for the analysis of this mode.

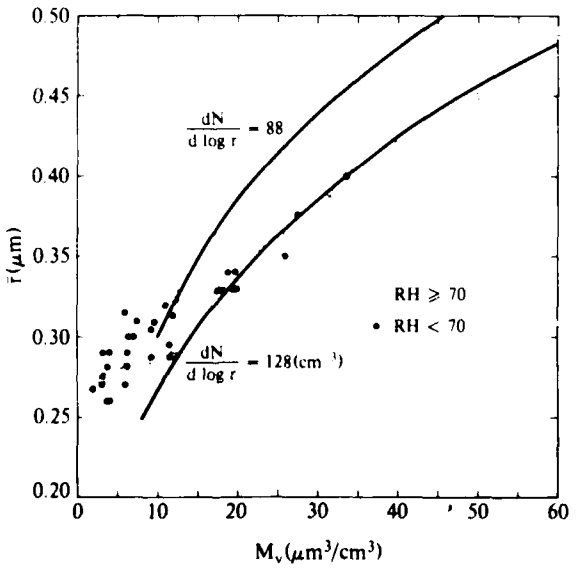

Fig. 3 The Mode Radius, $\bar{r}$, as a Function of the Maximum Volume, M_v, for all Data Collected within the Mixing Layer. Curves of $dV/dlogr$ for constant particle number density, $dN/dlogr$, (solid lines) show the $\bar{r}^3$ dependence of M_v. Data for which the relative humidity is greater than or equal to 70% are indicated by the open circles.

There does not appear to be a clear dependence on relative humidity as measured with a Cambridge Model 137-C3 aircraft hygrometer system. The open circles, representing cases where the relative humidity was greater than or equal to 70%, predominate at the larger radii as shown in Fig. 3. This apparent dependence on relative humidity, however, is mainly due to a small data sample taken at Rodby in an unusually heavy haze. If this Rodby data is neglected from the figure, then any tendency for the relative humidity to be greater than or equal to 70% when the value of the mode radius is greater than or equal to $0.34\mu m$ is probably not significant. There is a probability level of about 0.05 that the value of chi square will be equalled or exceeded by a random sample. If values greater than or equal to 80% are considered there again appears to be no clear dependence on relative humidity. This lack of a clear dependence on relative humidity may be a result of a drying of the aerosol in the intake plumbing.

In addition to measurements of the aerosol size distribution, the volume scattering coefficient, s, at $\lambda = 0.664\mu m$ was also measured. The scattering coefficient may be expressed as follows:

$$s = \pi \int_{logr_1}^{logr_2} Q_s(m,x) r^2 \frac{dN}{dlogr}\, dlogr$$

where $x = 2\pi r/\lambda$ is the size parameter of a spherical particle, $Q_s(m,x)$ is the efficiency factor for scattering (Van de Hulst, 1957), and r_1 and r_2 are the lower and upper limits of the particle radii under consideration. The relationship between s and the model parameters can be shown by using a log-normal description of the aerosol volume distribution and the expression $dV/dlogr = 4/3\ \pi\ r^3\ dN/dlogr$. After simple manipulation, s can be written in the form:

$$s = \frac{3}{4} \int_{logr_1}^{logr_2} Q_s(m,x)\ r^{-1}\ M_v\ e^{-0.5\left(\frac{logr - log\bar{r}}{\sigma}\right)^2}\, dlogr \qquad (1)$$

Even though the Royco is sensitive to a narrower particle size range than the nephelometer, calculations show that the distribution below $0.4\mu m$, typically, adds less than 15 percent to the total scattering at $\lambda = 0.455\mu m$. At the wavelength $0.664\mu m$ this percent contribution is even smaller, so the results of measurements by the Royco and

the nephelometer should, as seen in Eq. (1), be strongly related.

Values of the volume scattering coefficient, s, for the wavelength $0.664\mu m$ are plotted in Fig. 4 as a function of M_v. Though values of s are not uniquely dependent upon values of M_v, as seen in Eq. (1), the figure does show a trend of increasing s with increasing M_v. This trend can be explained by a shift in the volume distribution to larger radii for constant particle number, which increases M_v, and by increases in particle number which occur at the smaller values of M_v as shown in Fig. 3. The scatter in the data is, in part, a result of the fact that s was not always measured simultaneously with M_v. At times there was a two to three hour difference between the measurements. There is an apparent decrease in the scatter of the data when simultaneous measurements of s and M_v are plotted (circled data points). Some residual scatter can be expected due to the effects of variations in relative humidity which were detected by the nephelometer but probably not by the Royco due to drying in the intake plumbing.

In five of the flights studied, the air was so clear and the particle counts so low that there was no distinct accumulation mode measured at altitudes below the inversion height. All of these cases occurred during the winter season at Bruz, Rodby, and Meppen and, in all but one case, the airmass type was maritime. The average meteorological range, as estimated by the on-board observer, was always greater than 14 kilometers. The values of $dV/d\log r$ at $0.3\mu m$ were rarely greater than $2\ \mu m^3/cm^3$ (corresponding to a measured V over the range of the mode of less than $1\mu m^3/cm^3$) and s measured for the $0.664\mu m$ band was less than $6.8\times10^{-5} m^{-1}$. These values of $dV/d\log r$ and s are in agreement with the general trend of the data in Fig. 4, even though no value of M_v could be determined for these five cases.

To investigate the variation of M_v with respect to altitude, values of M_v were plotted against the ratio of the measurement altitude to the altitude of the inversion base. Data taken at Rodby on October 25 and 26 are presented in Fig. 5. In the mixing layer - an altitude ratio of less than 1.0 - the values of M_v decrease with increasing altitude for both days. For ratios greater than 1.0 (note the change of scale) the values of M_v are near zero.

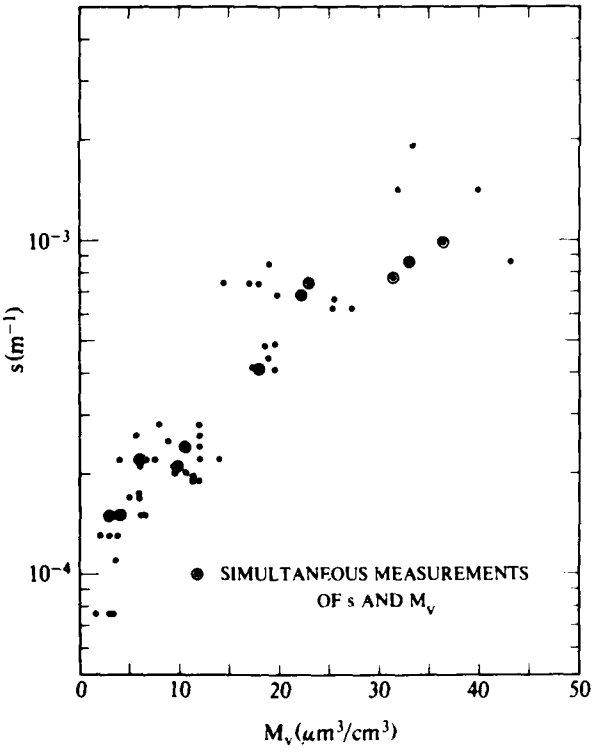

Fig. 4. The Volume Scattering Coefficient, s, for the Wavelength $0.664\mu m$ as a Function of the Maximum Volume, M_v. Cases where s and M_v were measured simultaneously are circled.

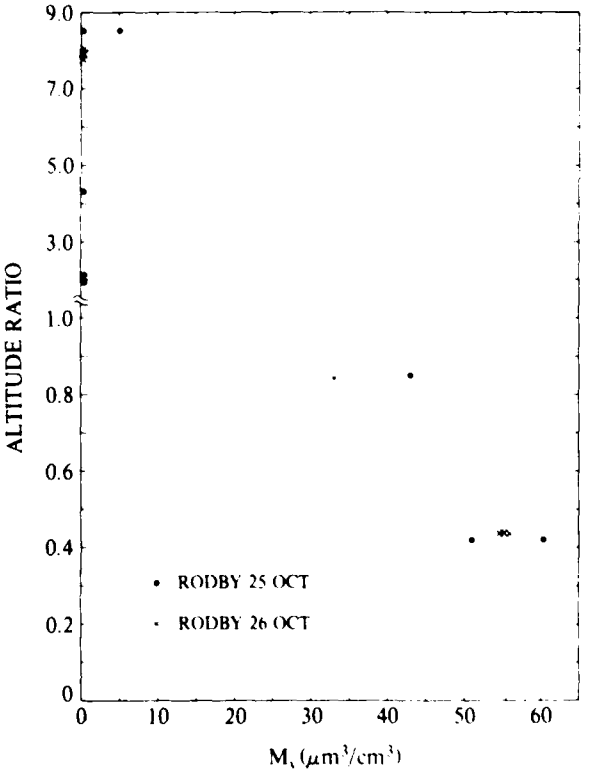

Fig. 5. The Maximum Volume in the Accumulation Mode, M_v, Measured at Rodby on October 25, 1976 (•) and on October 26, 1976 (x) as a Function of Altitude Ratio, the Ratio of the Aircraft Altitude during Measurement to the Thickness of the Mixing Layer.

All of the flights investigated show small values of M_v at altitude ratios of greater than 1.0. The decrease in M_v with increasing altitude in the mixing layer is, however, not always present. The results of measurements made at Bruz on July 4 and 7 are presented in Fig. 6. There is a decrease in the values of M_v with increasing altitude in the mixing layer for the July 7 data but not for the July 4 data, which appears invariant with altitude. The temperature profile to an altitude ratio of 0.4 shows July 7 as more thermally stable than July 4 at the time of the aerosol measurements. From the altitude 0.4 to 1.0 the temperature profile for both days was close to the dry adiabatic lapse rate. In addition, below the altitude ratio 0.4 the temperatures for July 4 were larger than those for July 7. This is probably because the July 7 data was taken in the morning and the July 4 data was collected in the afternoon. The scatter in the July 7 data at the low altitude ratios is in part a result of the time required to take these data, about 3.5 hours. In contrast, three measurements made at Bruz on July 6 over a 4 minute period at an altitude ratio of 0.3 show M_v ranging from only 15 to 18 $\mu m^3/cm^3$.

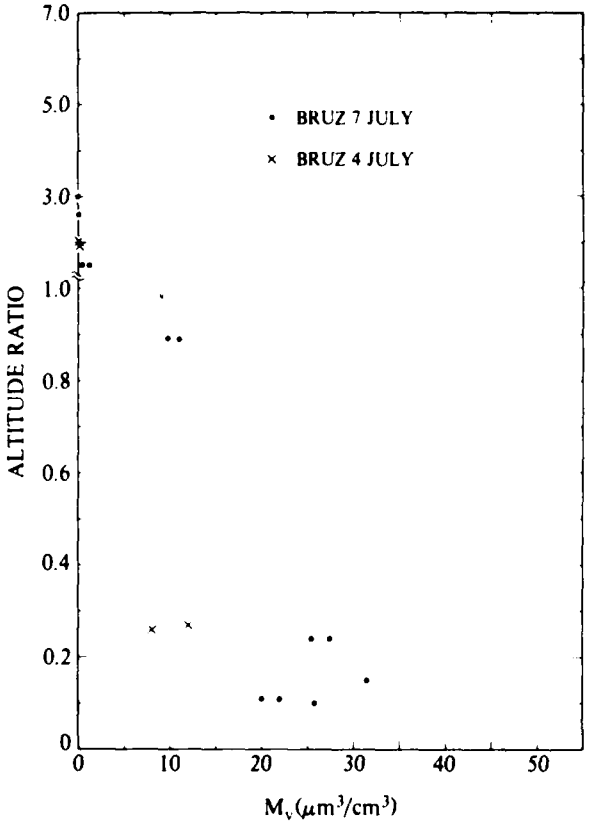

Fig. 6. The Maximum Volume in the Accumulation Mode, M_v, as a Function of Altitude Ratio for Data Collected at Bruz on July 7, 1977 (·) and on July 4, 1977 (x).

(b) COARSE PARTICLE MODE

Very often, low particle counts were measured in the size range of the coarse particle mode. These low counts made it difficult to determine values of the model parameters M_v, σ and $\bar{r}$ for this mode. Consequently, the coarse particle mode was represented by the integrated volume concentration, V, of particles from particle radius 2.0 to 5.7μm. A plot of V versus the altitude ratio for Bruz on July 4 and 7 is shown in Fig. 7. Within the mixing layer, the values of V decrease with increasing altitude for both days. Above the mixing layer, the increase in the values of volume concentration with increasing altitude ratio indicate the presence of a second haze layer. The on-board observer confirmed the existence of this second haze layer. It is interesting that there is no indication of a second haze layer in the accumulation mode data of Fig. 6. When there was no second haze layer above the mixing layer, the trend of V and M_v with altitude was similar.

The relationship between M_v of the accumulation mode and V of the coarse particle mode is represented in Fig. 8, which includes all the analyzed data from the mixing layer. The total volume concentration based on a log-normal fit to the accumulation mode can be

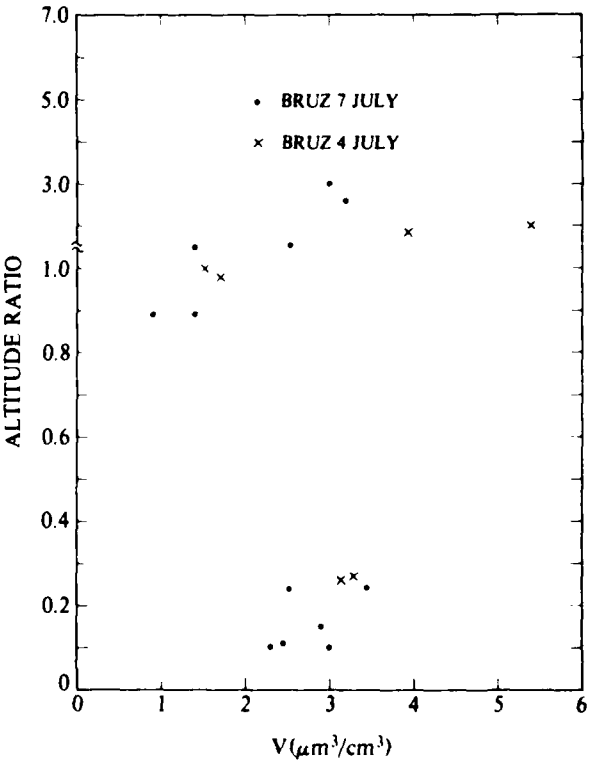

Fig. 7. The Volume Concentration of Particles in the Coarse Particle Mode, V, as a Function of Altitude Ratio for Data Collected at Bruz on July 7, 1977 (·) and on July 4, 1977 (x).

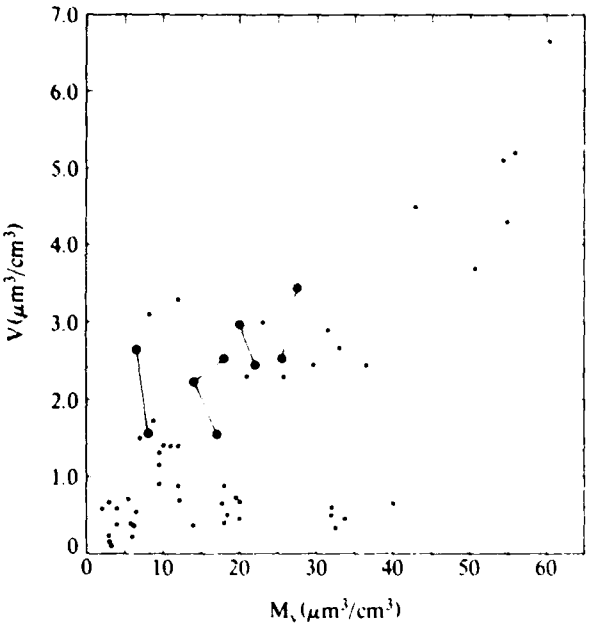

Fig. 8. The Volume Concentration of Particles in the Coarse Particle Mode, V, as a Function of Maximum Volume in the Accumulation Mode, M_v, for Data Collected within the Mixing Layer. The connected points show typical variations occurring over a period of 2 to 3 hours at a particular site and altitude.

determined by using the expression $V = M_v\,\bar{\sigma}(2\pi)^{1/2}$ where the average $\bar{\sigma}$ for the data set is 0.11. Values of volume concentration greater than 3.5 $\mu m^3/cm^3$ represent data from Rodby (October 25 and 26) taken in a saturated layer caused by the warm water of the Baltic and strong winds. The connected values in Fig. 8 show the variations that can occur over 2 to 3 hours at a given site and altitude. The set of data points occurring at M_v greater than 15 $\mu m^3/cm^3$ and V less than 1.0 $\mu m^3/cm^3$ are from two flights conducted on August 4 at the Meppen site. During the measurements, the air mass was classified as stagnant with ground stations in the area reporting haze, and winds less than 4 meters per second. The data in Fig. 8 indicate that values of M_v generally increase with values of V, except for the August 4 data. The increase is, however, strongly dependent on only two days of data from Rodby. It probably represents the effect of different air mass types on values of V and M_v and not the effect of a correlation between the two parameters. According to Whitby (1978) the two modes have different source and production mechanisms, so no strong correlation need exist between the two modes.

4. Discussion and Conclusions

Aircraft measurements of particle size distributions were made at several altitudes within and above the mixing layer at sites near Ahlhorn and Meppen (W. Germany), Rodby (Denmark) and Bruz (France). The experimental information gathered at the sites was analyzed in terms of volume-size distributions instead of the standard particle number distributions. The data support the following conclusions for the aerosol characteristics at the measurement sites:

1. There are at least two distinct modes comprising the size distribution. The accumulation mode is well defined by a log-normal distribution characterized by σ, $\bar{r}$, and M_v. Values of M_v appear to increase with values of $\bar{r}$. This is caused by an increase in particle number with increasing $\bar{r}$ for $\bar{r}$ less than 0.34μm, and it is a result of the $\bar{r}^3$ dependence of M_v. No clear relationship could be found between σ and the other model variables. An apparent relationship between M_v and the volume concentration of particles in the coarse particle mode, V, is probably the result of the different air mass types present during the study.
2. The values of M_v measured in the mixing layer appear well correlated with the measured values of the volume scattering coefficient.
3. The existence of an accumulation mode was confined to the mixing layer for the majority of the cases studied. During the winter season for conditions of good visibility in a maritime airmass, however, the particle counts were generally so low in the mixing layer that a distinct accumulation mode was not evident.
4. Within the mixing layer the value of M_v is not always constant with altitude, but may depend on the stability of the layer and the strength of the capping inversion.
5. Size distribution models for a specific air mass type which change only the total number of particles may not always be correct. Significant changes in $\bar{r}$ with changing M_v for a given air mass were observed. Work is currently underway to improve the determination of air mass type and to better understand its influence on the properties of the two modes.
6. The coarse particle mode was not confined to the mixing layer. It is interesting that haze layers above the mixing layer were found to have a distinct coarse particle mode but, generally, no accumulation mode. Its difficult to determine the origin of these layers which may be a specific source, or they may simply be aged aerosols.
7. The aerosol measurements probably represent a low humidity or dry particle distribution due to evaporation in the intake plumbing. Its likely that the shape of the distribution of each mode was preserved because of the small range in radius spanned by each of the two modes. Each of the modes may have been affected differently, however, due to differences in particle size and possible differences in chemical composition.

The rest of the aerosol data collected in northern Europe are being studied at present. With the added data

set, which is approximately twice as large as was used in this investigation, attention will be focused on how the characteristics of the accumulation and coarse particle modes change with air mass type and season. Techniques to better determine σ for the accumulation mode and M_c, r and σ for the coarse particle mode are being developed. In addition, the modeling of these modes at different altitudes in the lower troposphere based on a knowledge of the inversion height, volume scattering coefficient and air mass type is being pursued.

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